

# Fundamental Nuclear Data for Pinning down the Performance of Nuclear Weapons

Robert C. Haight, Mark B. Chadwick, and David J. Vieira

The cessation of nuclear-weapon testing and the new goal of predicting nuclearweapon performance from first principles have put increased importance on the accuracy of fundamental data on neutron-induced nuclear reactions. These data are crucial both for calculating the energy yield of a nuclear detonation and for benchmarking those predictions against the telltale radiochemical evidence recorded in the debris from past aboveground and underground nuclear tests. They may also be needed to interpret the nuclear activities of rogue nations and terrorist groups. In the past, many of the relevant reactions, especially those involving radioactive nuclei, have been too difficult to measure or have not been measured in the detail needed for modern weapons codes. As part of the Stockpile Stewardship Program, we have developed an unmatched suite of instruments capable of measuring neutron reactions on samples as small as a billionth of a gram. Together with the intense neutron sources at the Los Alamos Neutron Science Center and the Los Alamos National Laboratory's capabilities to produce isotopic samples, these unique instruments fill in much of the missing nuclear data and thereby reduce uncertainties in predictions of device performance.

On the opposite page: The intense flux of neutrons in a nuclear device causes several types of nuclear reactions, which are illustrated here. In fission, an incident neutron (blue) splits a fissile nucleus into two smaller nuclei, which release a number of neutrons, and those can induce more fissions. In neutron capture, a neutron is captured by a nucleus, changing it to a heavier isotope of the same element. The new isotope is left in an excited state, which de-excites to the ground, or lowest-energy, state by emitting a cascade of gamma rays (white). In the (n,2n) reaction, a neutron has high enough energy to knock out two neutrons, producing a lighter isotope of the same element.

n the sudden and gigantic release of energy by a thermonuclear weapon, two types of nuclear reactions are at work: the splitting of a very heavy fissile nucleus, typically uranium-235 or plutonium-239, and the fusion of two very light nuclei, deuterium and tritium. Both these reactions produce neutrons, and the exponential growth of the neutron population within a tiny fraction of a second, accompanied by an enormous release of energy, is the characteristic feature of a nuclear detonation. Predicting the history of neutron and energy production and the overall energy yield of a nuclear device from first principles is the goal of the Advanced Simulation and Computing (ASC) Program and of modern weapons codes. In this article, we outline the new and exciting approaches now being used at the Los Alamos Neutron Science Center (LANSCE) to achieve a much better understanding of the nuclear reactions that make weapons work and that give us insight into the performance of tested devices.

The importance of neutrons in nuclear weapons cannot be overstated. Neutrons induce fission, which is a major energy source in the explosion. Neutrons serve as the propagating vehicle for the supercritical chain reaction and the rapid, exponential amplification of energy. And neutrons can serve as essential diagnostic probes, either by escaping the device and entering external detectors or by transmuting materials placed in the device into radioactive nuclei. The product nuclei of the transmutations can be collected after the explosion and analyzed by radiochemical techniques to indicate the neutron flux in a specific region integrated over the history of the explosion (time-integrated flux is called "fluence"). Both these "prompt" and "radiochemical" (radchem) diagnostics give essential data that must be matched by calculations in order to benchmark the explo-

Table I. Neutron Reactions of Importance to Weapons Physics

Reaction	Droportion	Importance
Fission	Properties	Importance
1 1001011	Cross section	Reaction rate
	Energy release	Energy produced
		0, 1
	Neutron production	Neutron economy, <sup>a</sup> neutron output
	Gamma-ray production	Heating and gamma-ray output
	Fission products	Radiochemical diagnostics
	Fission products	Neutron absorbers or multipliers
Neutron capture		
	Cross section	Neutron economy
	Cross section	Radiochemical diagnostics
	Gamma-ray production	Heating and gamma-ray output
(n,2n)		
	Cross section	Radiochemical diagnostics
$(n,p), (n,\alpha)$ and others		
	Cross section	Radiochemical diagnostics

<sup>&</sup>lt;sup>a</sup> Neutron economy refers to reactions that can increase or decrease the number of neutrons in the system

sion codes. To interpret these data correctly, one must know the basic nuclear transport and reaction data with high accuracy.

The need for improved understanding of basic nuclear reactions increased greatly with the cessation of underground nuclear tests in 1992. Whereas before that date weapons could be tested and a large database was accumulated of systematic performance, now the performance of a weapon is known only through calculation. These calculations are adjusted to reproduce previous test data, but their relevance as benchmarks can be questioned when conditions change because of aging materials, component modernization, or new designs. Our explosion models could be inappropriate for simulating completely different weapon concepts that might

be used by rogue nations or terrorists. Thus, it is critical to improve the models so that their application in new areas will give credible and accurate results. If the input neutron-reaction data are inaccurate, then the predictions of the models certainly will be so, and because of the high nonlinearity of the physical processes in a nuclear explosion, the models could be grossly in error. Thus, one of the most basic requirements in understanding previous test data to improve the models is highly accurate neutron-reaction data.

With the rapidly increasing power of computers, the actual nuclear properties can be used in modern weapons codes, rather than some convenient (and somewhat inaccurate) approximation to the data. A further increased need for nuclear data has been added

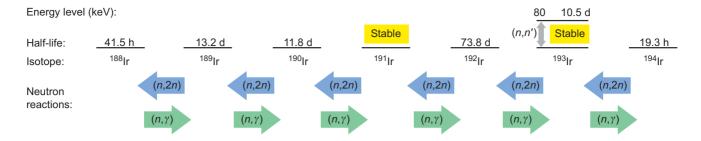


Figure 1. Chain of Neutron-Induced Reactions on Iridium Used in Radiochemical Analysis Neutron-induced reactions on iridium isotopes are used to infer neutron fluence by radiochemical analysis of the amount of the radioactive isotopes produced. Natural iridium consisting of the stable isotopes 191 and 193 was placed in the device. The radioactive isotopes produced in the test live long enough so that they can be recovered by drill-back procedures and then analyzed in the laboratory. The (n,2n) reactions have thresholds of approximately 6 MeV and therefore indicate the fluence of neutrons above this energy. The  $(n,\gamma)$  reactions have zero threshold and indicate the neutron fluence generally below 1 MeV. The (n,n') reaction on iridium-193 producing the 10.5-day excited state at 80 keV is sensitive to neutrons between 1 and 5 MeV, which is the major energy range for neutrons produced in fission.

recently as designers are now asked for uncertainties in the predicted performance. Quantification of margins and uncertainties (QMU) is now central to the weapons program. Fortunately, data from previous nuclear tests include extremely careful measurements of total energy yields, neutron and gamma-ray output, and the production of radioisotopes, which were extracted from weapon debris with radiochemical techniques. To interpret these data, we need precise fundamental nuclear data, rather than systematics of questionable accuracy, as input to these codes. The important nuclear reactions that must be quantified as accurately as possible are listed in Table I.

Nuclear explosions are unique among phenomena that happen on earth in that they obviously occur extremely fast and have a very large number of neutrons in a small volume driving up the rate of nuclear reactions. Only some astrophysical sites have similar conditions. With these special conditions of short time scales and very high reaction rates, nuclear reactions that are not important in systems such as nuclear reactors become very important in nuclear weapons. For example, new nuclei with short half-lives can be produced in the

explosion and then undergo additional reactions themselves before the explosion is over. In other cases, nuclei can be excited to states that live longer than the explosion, and these so-called "isomeric" states can have properties different from those of the ground state. In both cases, new opportunities arise to understand the performance of a nuclear weapon. Along with these opportunities, new challenges present themselves and require new, morepowerful techniques to take advantage of the opportunities.

Our nuclear-data research program at LANSCE has two components. One is to measure much more accurately reactions on stable (or at least longlived) nuclei used as fuels, structural materials, or radiochemical detectors. Although many of these materials have been studied before, there are often large discrepancies in the literature that need to be resolved. And there are also many gaps in the literature data. The other component is a completely new challenge, namely, to study reactions on radioactive targets that have heretofore been impossible because of the short half-lives of the nuclei or the difficulty of producing or using these highly radioactive materials.

An example of basic nuclear

reactions used to infer the neutronic performance of a nuclear weapon is given in Figure 1. If iridium, a naturally occurring element that has two stable isotopes, is placed in the device, radioactive isotopes and longlived excited nuclear states (isomers, for example, iridium-193m) are created by the intense neutron flux of the explosion. In the debris recovered in drill-backs hours or days later, these radioactive products can be separated and analyzed to indicate the neutron fluence and its energy spectrum at the location of the loaded iridium. Knowledge of the individual reaction rates is essential to the analysis, and our challenge is to improve this knowledge in key areas that have heretofore been inaccessible.

## LANSCE, a Powerful Source of Neutrons

In the past, acquiring accurate nuclear-reaction data was limited by the lack of suitably intense neutron sources. To address the present-day requirement for more-accurate nuclear-reaction data, an advanced neutron source is essential. LANSCE is arguably the best facility in the

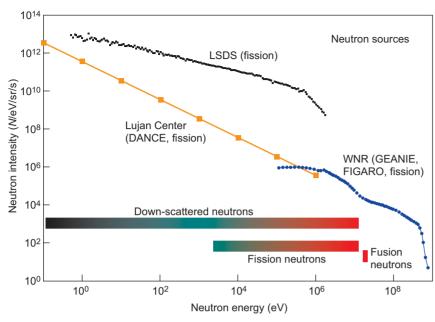


Figure 2. Energy Ranges of Neutron Sources at LANSCE
The intensity and energy distributions of the neutrons produced at the WNR,
Lujan Center, and LSDS show the very large range of energies covered in these
facilities.

world to fill in the missing pieces of this nuclear puzzle. LANSCE is centered on a powerful 800-MeV proton linear accelerator used to produce intense neutron beams. The facility consists, in fact, of three neutron sources used for nuclear physics for the weapons program:

• The Weapons Neutron Research (WNR) source of fast neutrons from 100 kilo-electron volts (keV) to 800

million-electron volts (MeV) in energy
• The Lujan Neutron Scattering Center

- The Lujan Neutron Scattering Center (Lujan Center) source of moderated neutrons from subthermal energies to 500 keV
- The Lead Slowing-Down Spectrometer (LSDS) for specialized experiments with neutron energies from 1 electron volt to 200 keV

The relationship of the neutron ranges spanned by these three facili-

ties to the energy range of interest for weapons is shown in Figure 2. LANSCE produces the most intense source of neutrons in the world over the range of energies relevant to weapons. Future improvements are expected to increase the usable neutron flux in the important weapon regime of 1 keV to 5 MeV by a factor of 30 or so by stacking pulses in the Proton Storage Ring at LANSCE and transporting the stacked pulses to the WNR. At all these sources, neutron reactions are studied over a large range of energies all at once, using the time-of-flight method described below.

At LANSCE, neutrons are produced by spallation, which means that each time a proton pulse hits a heavy metal target, a pulse of neutrons having a wide range of energies boils off, or evaporates, from the target nuclei. Time-of-flight techniques are used to indicate the energy of the incident neutron. For the WNR and Lujan Center sources, the sample under study and the measuring instruments are placed at some distance (9 to 90 meters) from the source. Because the source is pulsed, neutrons with higher energy reach the measuring station before the slower neutrons (Figure 3), and the neutron time of flight can be measured to determine the incident-neutron energy. At the

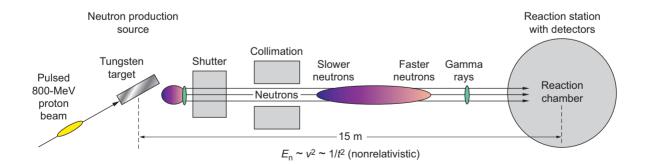


Figure 3. Determining Neutron Energies by Time-of-Flight Techniques
Neutron energies initiating the reaction are determined by their time of flight (t) from the neutron source to the reaction station. The source is pulsed and the more-energetic (faster) neutrons arrive at the reaction station before those with lower energy, which travel more slowly.

LSDS, similar techniques are used although there is not a defined flight path (see below). This ability to study a given reaction simultaneously over a large energy range provides a consistent set of data over this range and therefore much more accurate information for the database.

### **Innovative Instruments** at LANSCE

The second essential requirement in improved experimental capabilities is to be able to take advantage of these intense neutron sources. The challenge is to develop instruments that are sensitive to the desired signal and are not overwhelmed by high count rates or by backgrounds from unwanted sources. In this article, we describe unique high-precision experiments performed with several new LANSCE instruments-known as GEANIE, DANCE, FIGARO, and LSDS—to fill in significant gaps in the nuclear data. We outline several measurements to improve the interpretation of radchem results and also describe developments that are reducing uncertainties in the prediction of fission performance.

## **GEANIE**—Reactions to Determine Neutron Fluence in Data from Weapon Tests

GEANIE (Germanium Array for Neutron-Induced Excitations) is an array of 26 high-resolution gamma- and x-ray detectors that view a sample bombarded with neutrons with energies from 0.1 to 200 MeV from the WNR source of fast neutrons (Figure 4). This unique array was constructed by a collaboration of researchers from Los Alamos and Lawrence Livermore National Laboratories specifically to investigate neutron reactions at the center of rad-



Figure 4. The GEANIE Detector
GEANIE is composed of 26 high-purity germanium detectors. The photo shows
them viewing gamma rays from an encapsulated plutonium-239 sample bombarded with neutrons from the WNR source.

chem diagnostics, reactions that were tantalizingly difficult to study by any other means. The next two sections describe two of these experimental challenges: One is used to interpret radchem data for the neutron fluence in a weapon above 6 MeV, and the other is a unique probe into the fission neutron spectrum in the 1- to 5-MeV range.

## First Accurate Data to Interpret Plutonium Radchem Results.

Isotopes produced during an explosion in the nuclear fuels, whether

uranium or plutonium, serve as a very useful radchem diagnostic of weapon performance. A neutron impacting a plutonium-239 nucleus can cause fission, but if the energy of the neutron is greater than 6 MeV, it can alternatively induce an (n,2n) reaction that transforms plutonium-239 to plutonium-238. We write this reaction as  $^{239}$ Pu $(n,2n)^{238}$ Pu. Thus, a measurement of the amount of plutonium-238 provides valuable information on the neutron energies in the device above 6 MeV, provided one knows the cross section for producing plutonium-238

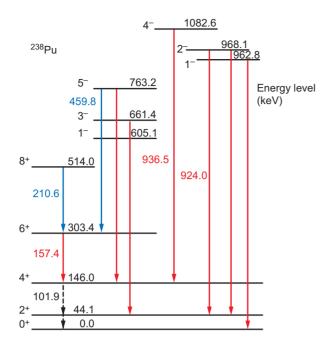


Figure 5. Gamma-Ray Transitions Observed in the  $^{239}$ Pu $(n,2n)^{238}$ Pu Reaction

The yields of the transitions shown in red were used to deduce the total cross section for this reaction with good confidence.

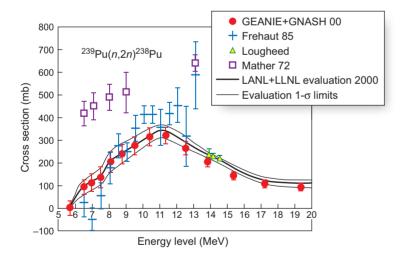


Figure 6. Measurement Results of *n*,2*n* on Plutonium at GEANIE vs Previous Data

The  $^{239}$ Pu(n,2n) $^{238}$ Pu cross section results deduced from the GEANIE measurements are compared with previous data. Note that this cross section has a threshold at 6 MeV, and so the amount of plutonium-238 produced in a nuclear-weapon test is sensitive to neutrons above this energy.

as a function of energy.

Surprisingly, the first accurate measurements of the cross section ("rate") for the  $^{239}$ Pu $(n,2n)^{238}$ Pu reaction over the neutron energy range of 6 to 20 MeV were not feasible until only a few years ago, when the new GEANIE detector at the WNR came online. The two previous approaches turned out to have insurmountable difficulties to yield anything that was close to an accurate result over the energy range of interest.

One would think that this reaction could be studied in the laboratory by bombarding a sample of plutonium-239 with monoenergetic neutrons and then measuring the amount of plutonium-238 produced. However, there is the very real problem that even the purest sample of plutonium-239 has some plutonium-238 contaminant. To overcome that background, one needs a very large total number of incident neutrons so that the buildup of plutonium-238 from the reaction is large enough to measure with radiochemical techniques. This direct approach was pursued many years ago with the intense 14-MeV neutron source at Lawrence Livermore. However, at other neutron energies, especially in the threshold region near 6 MeV, there has simply not been any neutron source intense enough for this measurement.

The other approach used previously was to detect the two neutrons emitted in the (n,2n) reaction, but here another difficulty frustrated the researchers. The incident neutrons sometimes induce fission of plutonium-239 that is accompanied by the release of two neutrons, and those fission neutrons look just like the two neutrons coming from the (n,2n) reaction. In fact, the fission neutrons are sufficiently numerous to introduce large uncertainties in the deduced cross section for the reaction 239Pu $(n,2n)^{238}$ Pu.

In 1996, the GEANIE detector

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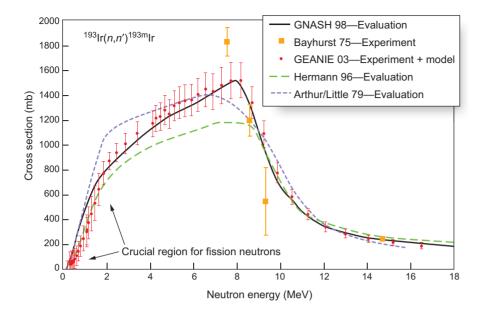


Figure 7. New Evaluated Production Cross Section for Iridium-193m The new GEANIE/GNASH result for the 82-keV isomer production cross section in iridium-193 is shown here covering the incident-neutron energy range from the reaction threshold (80 keV) to 18 MeV. The 1- $\sigma$  standard deviations come from uncertainties in both GEANIE data and GNASH reaction modeling. This new cross section is compared with the historical one by Edward Arthur and Robert Little (of the Theoretical and Applied Physics Divisions at Los Alamos), which was previously used at the Laboratory, with a more recent calculation by GNASH in 1998 and with another evaluation. The previous Los Alamos data by Bayhurst are also shown.

was completed at LANSCE, and its availability opened up a totally new approach. The plutonium-238 nucleus produced in the  $^{239}$ Pu $(n,2n)^{238}$ Pu reaction is most likely left in an excited state that quickly emits gamma rays as it decays to the ground state. In other words, the (n,2n) reaction should be written as  $^{239}$ Pu(n,2n +gammas)<sup>238</sup>Pu. The GEANIE detector is an ideal instrument for measuring those gamma rays with high-energy resolution and high efficiency. The nuclear energy-level diagram in Figure 5 shows the transitions in the plutonium-238 nucleus that produce the gamma rays. Following the measurements at GEANIE carried out by a team from Lawrence Livermore and Los Alamos, model calculations were conducted in the Theoretical Division

at Los Alamos to relate the measured production of specific gamma rays produced in the (n,2n) reaction cross section to the total production of plutonium-238. The larger the fraction of gamma rays detected by GEANIE, the better constrained is the theoretical model, and the lower is the resulting uncertainty in the deduced cross section. The results are shown in Figure 6, along with the previous Lawrence Livermore data at 14 MeV described above, the rather imprecise data from measuring the two neutrons, and a wide range of estimates of the cross section made before the GEANIE experiment.

The final results from this GEANIE project are crucial to inferring details of weapon performance through analysis of the measured plutonium-238/plutonium-239 isotopic ratio in the debris of the explosion. This GEANIE measurement earned a National Nuclear Security Administration Award of Excellence for the Los Alamos and Lawrence Livermore collaboration.

First Accurate Data to Diagnose **Fission Neutron Flux from** Radchem Results. In a more recent study, the GEANIE detector was used to determine the cross section for another important radchem reaction—the inelastic neutron-scattering reaction  $^{193}$ Ir(n,n') $^{193}$ mIr, in which the incident neutron excites the iridium-193 nucleus to a so-called "isomeric excited state" with a halflife of 10.5 days. This isomer-producing reaction is particularly interesting because, unlike the (n,2n) radchem detectors, such as  $^{239}$ Pu $(n,2n)^{238}$ Pu, which are sensitive to high-energy neutrons (above 6 MeV), or the neutron capture detectors, which are sensitive to low-energy neutrons (below 1 MeV), isomer production via (n,n')reactions is sensitive to neutrons in the intermediate energy range of 1 to 5 MeV. Neutrons released in fission typically have energies of 1 to 5 MeV, and thus isomer production is a good measure of fission neutrons.

Surprisingly, accurate experimental data for the isomer production cross section were lacking for many years, and weapon designers had to rely almost completely on theory to interpret radchem results for iridium. Unfortunately, the predictions of the theory were acknowledged to be very imprecise because of poorly known nuclear-structure details in the (n,n') nuclear reaction. That situation has changed through the latest measurements at GEANIE.

In this case, as in the cross section measurements discussed in the previous section, direct laboratory experiments to measure the isomer production proved extremely difficult

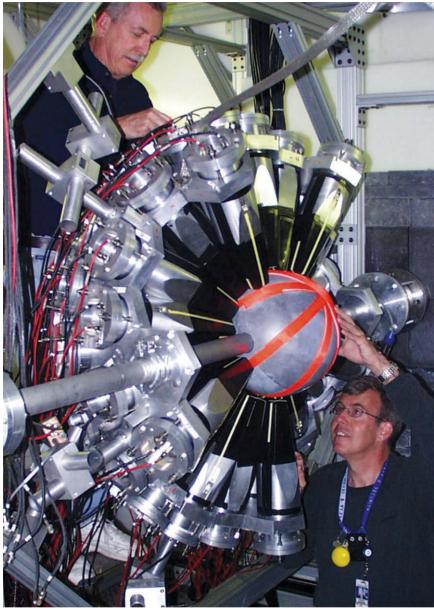


Figure 8. DANCE
A photo of the DANCE detector shows only one-half of the array of barium fluoride (BaF<sub>2</sub>) gamma-ray detectors and the spherical lithium hydride (<sup>6</sup>LiH) neutron absorber surrounding the sample. When in use, the other half of the detector array is moved into place to make a spherical shell of gamma-ray detectors.

and gave results with huge error bars (Figure 7). The source of this difficulty was that the isomer decays mostly by a low-energy electron and low-energy x-rays, both of which are difficult to measure if only small quantities of the isomer are present.

Previously, there was only one set of data with large uncertainties at neutron energies of 7.6, 8.6, 9.3 and 14.7 MeV, and none in the region of most interest from 1 to 5 MeV.

As in the (n,2n) measurements on plutonium, the advent of GEANIE

opened up an indirect approach to measuring isomer production, namely, through measuring the gamma rays emitted in the decay of excited states of iridium-193 to the isomer. The GEANIE/GNASH results resolved the long-standing question of what shape the cross section has as a function of incident-neutron energy, especially in the 1- to 5-MeV region that is so important for diagnosing fission neutrons.

#### DANCE: Unraveling Contributions from Complicating Reactions

DANCE, the Detector for Advanced Neutron-Capture Experiments (Figure 8), is shaped like a soccer ball with 160 equal-area pentagonal and hexagonal elements made from 15-centimeter-thick barium fluoride (BaF<sub>2</sub>) scintillation material to capture nearly all the energy of gamma rays emitted in a nuclear reaction. Because of its high segmentation, the multiplicity (or number) of gamma rays in the reaction can also be measured. The sample is placed in the center of this spherical array and is bombarded with neutrons from the Lujan Center neutron source covering the energy range from thermal to approximately 200 keV. The most likely reaction on typical materials at these energies is neutron capture, whereby the neutron is added to the target isotope increasing its mass by one unit. The energy liberated in the capture process appears as one or more (typically 3 to 5) gamma rays. In this reaction, the total energy of all the gamma rays is characteristic of the sample under study, and this total energy is between 4 and 9 MeV. Thus, by energy selection and by the number of gamma rays detected, DANCE has good specificity for defining the particular neutron-capture reaction of interest. It also minimizes the back-

ground from scattered neutrons, as we have placed a lithium hydride (<sup>6</sup>LiH) shell around the sample to absorb these neutrons. Because this detector is so efficient and the neutron flux at the Lujan Center is so high, sample sizes can be very small—only about 1 milligram. With such small samples possible, a new world of reaction studies with radioactive samples is now open to investigation.

The DANCE instrument was developed to investigate reactions complementary to those studied at GEANIE. It is designed to quantify reactions that can confuse the interpretations of radchem results, such as from the neutron reactions just mentioned, by addressing the neutron energy range below 200 keV. The ultimate fate of weapon neutrons is often determined in this energy range because, once produced, these neutrons lose energy by reactions and scattering. Since radchem diagnostics are time integrated, the reactions produced by these slow neutrons must be taken into account in interpreting the test data correctly.

#### **Neutron Capture Reactions.**

Reactions used for weapon diagnostics such as those described above,  ${}^{239}$ Pu $(n,2n)^{238}$ Pu, and  $^{193}$ Ir(n,n') $^{193}$ mIr, produce radioactive products that can be assayed by radiochemical techniques. Many other reactions have also been used in nuclear tests, and the amount of each of the radioactive isotopes produced indicates both the fluence and the energy distribution of the neutrons at the location of the initial isotope. Now suppose that the radioactive isotope produced in the test undergoes further nuclear reactions in the dense neutron environment. Then the abundance of the radioactive isotope will be changed by second-order reactions, such as neutron capture reactions. These effects can provide useful information about the number

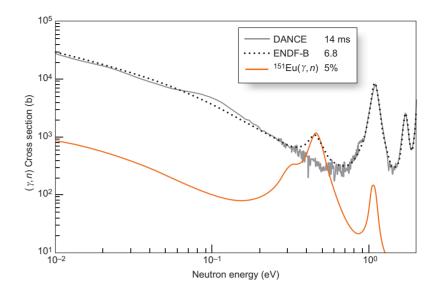


Figure 9. Neutron-Capture Cross Section of Samarium-151 DANCE data on the neutron-capture cross section on the radioactive nucleus samarium-151 show that the existing ENDF<sup>1</sup> evaluation incorrectly puts a resonance at 0.46 eV. This evaluation was guided by an experiment done elsewhere with a sample that was contaminated by a 5% impurity of europium-151, the decay product of samarium-151.

of low-energy neutrons, but they can also change the relative amounts of isotopes produced via (n,2n) or (n,n') reactions in a weapon test and thereby complicate the analysis. Here follows an example based on the element thulium, in which both processes occur.

The rare-earth element thulium has been used as a neutron fluence detector in weapon tests. It has only one stable isotope, thulium-169, which, under intense neutron bombardment, makes a whole series of radioactive isotopes of the element, from masses 167 to 172. The chain of reactions is similar to that in Figure 1. The capture reaction  $^{169}\text{Tm}(n,\gamma)^{170}\text{Tm}$  gives data on low neutron energies, below 1 MeV, and we are remeasuring this cross section in order to resolve discrepancies between previous

measurements. The (n,2n) reactions, especially  $^{169}\text{Tm}(n,2n)^{168}\text{Tm}$  and  $^{168}$ Tm $(n,2n)^{167}$ Tm, probe neutron energies above 6 MeV. The ratio of the amounts of two radioactive isotopes, such as thulium-167 and thulium-168, is often considered a reliable diagnostic of the neutron fluence above 6 MeV because isotopes of the same element undergo the same chemistry and fractionation in the underground environment. Although it is very difficult to measure the (n,2n) reaction on the radioactive thulium-168 nucleus, systematics of nuclear reactions, together with a heroic point at 14 MeV from Livermore, can be used here rather reliably to estimate the cross section as a function of energy for this reaction. So, barring any complications,

<sup>&</sup>lt;sup>1</sup> Evaluated nuclear data result from analyzing all available experiments, resolving discrepancies, and determining both the values and the uncertainties. They are kept in libraries known as ENDF, the Evaluated Nuclear Data File. ENDF data are freely available from the National Nuclear Data Center at Brookhaven National Laboratory (http://www.nndc.bnl.gov) and from the Los Alamos Web site (http://t2.lanl.gov).

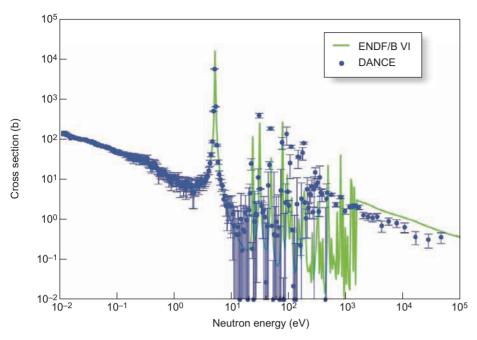


Figure 10. Neutron-Capture Cross Section of Uranium-234
The important region above 1000 eV was never been measured before with good resolution. The data obtained at DANCE indicate that the evaluated data accepted for use in the United States, the ENDF/B-VI values, are significantly too high in this energy region.

the ratio of thulium-167 to thulium-168 gives good information on the neutron fluence above 6 MeV. Now, suppose, however, that another, second-order reaction comes into play, the  $^{168}\text{Tm}(n,\gamma)^{169}\text{Tm}$  reaction, which decreases the amount of the originally produced thulium-168. The cross section for that reaction is not at all well known. Theory is not of much help because, where we have data, theory predicts capture rates only to a factor of 2 for stable nuclei. (However, more-precise predictions are sometimes feasible where nuclear modeling input parameters for a specific reaction are known from other nuclear physics studies.) In this neutron-capture reaction, the target nucleus thulium-168 is unstable, and there are no tests of the theory in this mass range for unstable nuclei. Similar issues come into play for the  ${}^{170}\text{Tm}(n,\gamma){}^{171}\text{Tm}$  reaction. For these reactions, experimental data are essential, and that is why DANCE

was constructed.

A series of radioactive isotopes of importance to radiochemical diagnostics is being prepared for experiments at DANCE. In preparation for this campaign, we have studied the available isotope samarium-151, which is of interest to astrophysicists in understanding the formation of elements in stars. These data are shown in Figure 9. Previous estimates were confirmed above 1 electron volt, and a significant correction was found near 0.46 electron volt.

DANCE is not limited to rareearth nuclides and has already produced data on neutron capture for the important set of uranium isotopes uranium-234, -235, -236, and -238 and for neptunium-237. Neutron-capture data for uranium-234 are shown in Figure 10. These data are the first reliable ones for incident-neutron energies above a few electron volts. In a weapon environment, neutron reactions on the longer-lived isotope uranium-235 create shorter-lived isotopes, which can subsequently participate in neutron reactions. Understanding neutron-capture cross sections on uranium-234 and -236 will improve the analysis of the radio-nuclide debris data after an explosion.

More generally, DANCE brings the unique capability to reduce many of the uncertainties that have been associated with nuclear reactions on short-lived isotopes, making a contribution crucial for an improved understanding of device performance.

With only very small samples required, DANCE has made it feasible to acquire adequate data on short-lived radioactive nuclei and rare nuclei. Reactions on these rare species are of interest beyond the nuclear-weapon community because they also occur in the interior of stars. In fact, there is now underway at LANSCE an exciting project to study neutron capture reactions that will help explain the dynamics of red giant stars and their synthesis of the heavy elements through slow neutron capture (see the sidebar "Stardust and the Secrets of Heavy-Element Production" on page 70).

#### FIGARO: Mapping the Energy Dependence of Neutron Emission

FIGARO (Fast-Neutron-Induced Gamma-Ray Observer) is an array of 20 neutron detectors and two or three gamma-ray detectors (Figure 11), all of which look at a sample bombarded with fast neutrons (0.1–200 MeV) at the WNR neutron source. The gamma-ray detectors are rather close (15 centimeters) to the sample, but the neutron detectors are set back 1 to 2 meters from the sample in order to determine the outgoing neutron energy by time of flight from the sample. Two types of samples are used: active samples, such as fission chambers that produce a signal when

a nuclear interaction has taken place, and passive samples, which do not produce any signal. For the latter, gamma rays emitted in the reaction and detected by the gamma-ray detectors indicate the occurrence of the reaction. We need one or the other of these signals to measure the time of flight of the incident neutron from the source to the sample to deduce the incident-neutron energy of the event. We need the same signal as a "start" signal for time of flight of the neutrons emitted in the reaction from the sample to the neutron detectors. This latter time of flight gives the energy of the outgoing neutron. We call this setup a "double time-offlight experiment," which has been attempted at other laboratories but is now much easier to accomplish because of the intensity and pulse structure of the WNR neutron source. With this capability, we are able to map out the dependence of the neutron emission spectrum as a function of incident-neutron energy, all in one experimental run.

The importance of these neutron emission spectra is clear: Neutrons are the vector for nuclear chain reactions in a nuclear detonation. That is, when a neutron-induced fission takes place, more neutrons are emitted, and these can then induce further fissions. It is well known that both the number and energy spectrum of the emitted neutrons depend on the energy of the neutron inducing the fission. A model developed at Los Alamos (referred to as "the Los Alamos model") for this spectrum is well known and used worldwide. Although the model has been tested several times at specific energies, it still needs to be validated for the whole range of incident-neutron energies for which it was originally intended (incident energies less than 20 MeV) and for extensions to higher incident energies. FIGARO enables comprehensive studies of these fission properties.



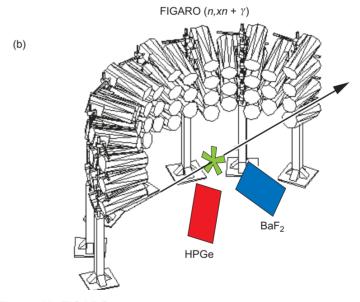


Figure 11. FIGARO

FIGARO, the Fast-Neutron-Induced Gamma Ray Observer at the WNR, is designed to measure neutron-induced reactions that result in the emission of neutrons and gamma rays. (a) The photograph shows the liquid scintillation neutron detectors, the fission chamber, and the location of the neutron beam. (b) The diagram shows a carefully collimated beam hitting the sample, which can be a fission chamber containing fissile material. The energies of the incident neutrons are determined by time of flight from the neutron source to the sample. Similarly, the energies of the fission neutrons are measured by time of flight from the sample to the array of neutron detectors (currently, there are 20 detectors) located 1 to 2 m from the sample.

The other application of FIGARO is to neutron emission in reactions on nonfissile nuclei such as structural materials. In many tests of nuclear weapons, the neutron spectrum emitted from the device was measured, and here, of course, the neutrons needed to penetrate through structural materials. These interactions need to be included in the calculations in order to relate the measured spectrum to that inside the device.

## First Complete Data for Energy Spectrum of Uranium Fission

Neutrons. Together with scientists from the Commissariat à l'Énergie Atomique (CEA) at Bruyères-le-Châtel in France, we are using the FIGARO array of neutron detectors to measure the fission-neutron energy spectrum and the average neutron multiplicity for neutron-induced fission. Our first experiments were on uranium-238 and -235 for incidentneutron energies from 1 to 200 MeV. The results from 1 to 20 MeV are important for weapon applications, whereas those for higher energies are relevant to accelerator-driven systems or for irradiation facilities such as the Materials Test Station (MTS) to test the radiation effects in materials for the Advanced Fuel Cycle Initiative (AFCI)—see the article "The Role of LANSCE in the Nuclear Energy Future" on page 124. Measurements in progress are on neptunium-237. It is staggering that, to date, there are only very sparse data for the promptfission neutron energy spectrum of plutonium (a more difficult sample) for incident-neutron energies above a few million-electron volts. Such a measurement is urgently needed and will be carried out at FIGARO in the near future.

The average energy of the fission neutrons determined in this way shows the general trend and an essential physics feature (Figure 12). For uranium-238, for example, one

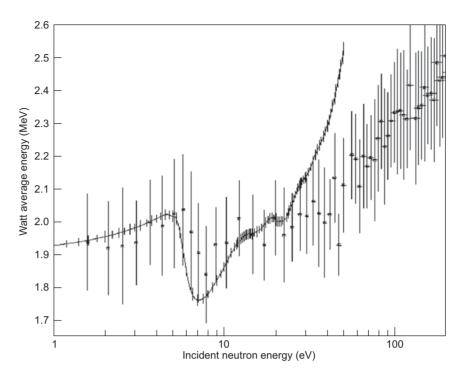


Figure 12. Average Energy of Fission Neutrons from Uranium-238 The average energy of fission neutrons from uranium-238 bombarded with neutrons was measured by FIGARO in the incident-neutron energy range of 1 to 200 MeV. Note the inflexion of the data and the model (solid curve) near 7 MeV, the energy at which "second-chance" fission begins.

sees a gradual increase in the average energy with incident-neutron energy except near 6 MeV, at which value a second fission channel opens up. That channel is the reaction  $^{238}U(n,n)$ + f) channel, or "second-chance fission," in which a neutron is emitted before fission and carries off a significant amount of binding energy. These second-chance fission neutrons contribute to the chain reaction and thus can impact weapon performance. The energy distribution of the fission neutrons needs to be included in the calculations, as these neutrons are responsible for continuing the chain reaction. Our data validated very nicely the Los Alamos model in the region of importance to weapons, that is, below an incident-neutron energy of 20 MeV. At higher energies, which might be of importance to accelerator-driven fission systems, the Los

Alamos approach as implemented by our CEA colleagues does not do so well at predicting the average neutron energy.

Although the average number of neutrons emitted in each fission event is rather well known for very low incident-neutron energies, a complete mapping including the angular distribution of these neutrons has never been done, and the average number of emitted neutrons is not known for fission induced by high-energy neutrons. We have completed the multiplicity measurement, which showed interesting differences between the uranium-235 and -238 target nuclei (Ethvignot et al. 2005). Angular distribution information requires longer experimental runs, which have been carried out, and the data are now being analyzed. All of these data pertain to the basic understanding of the

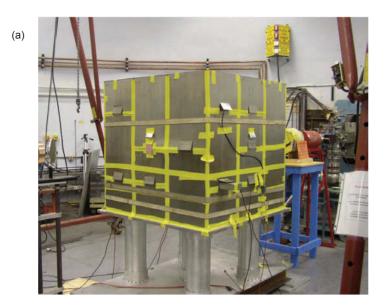
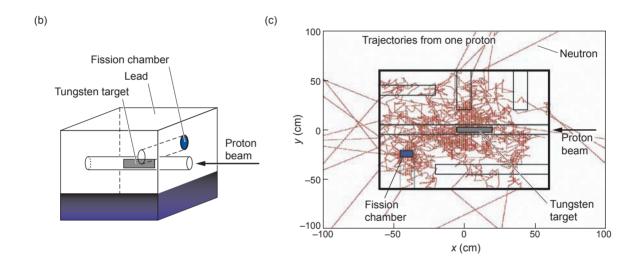


Figure 13. The LSDS (a) The LSDS, the Lead Slowing-Down Spectrometer, is composed of 36 lead blocks. which are assembled into a cube 1.2 m on a side. (b) A pulsed proton beam from the LANSCE Proton Storage Ring hits a tungsten target in the center of the cube to produce a large number of neutrons, which then slow down by scattering from the lead nuclei. (c) The red trajectories, calculated with a Monte Carlo code, are an example of neutron trajectories resulting from one proton incident on the tungsten. A pulse of  $1.5 \times 10^{11}$  protons produces trajectories that fill the entire cube. Detectors are placed inside the channels, and reactions are recorded as a function of time after the proton pulse. The neutron energy inducing a reaction is directly related to this time interval.



fission process, which is still a very active area of research worldwide. Open questions include the following: (1) How does a nucleus undergo fission when it is highly excited? (2) For excited nuclei, how many neutrons are emitted before fission occurs? (3) When an energetic neutron interacts with a nucleus, does the system emit neutrons before coming to a state in thermal equilibrium, which then fissions? (4) What are the channels through which fission

occurs in terms of the time-dependent elongation, deformation, mass asymmetry, and other semiclassical shape parameters?

FIGARO is still in the developmental stage, and we plan to add additional detectors to increase the efficiency of measuring coincidences of fission events and fission neutrons, as well as coincidences of incident neutrons and emitted gamma rays. The latter are important for fundamental studies of the excited states of nuclei.

#### LSDS: Studying Fission Reactions on Ultrasmall Samples

The LSDS (Figure 13) is a unique instrument consisting of 36 very large lead bricks assembled into a 20-ton cube, 1.2 meters on a side, with a tungsten cylinder in the center and a path for the incident protons to hit the tungsten directly. Bunched and pulsed 800-MeV protons from the Proton Storage Ring (PSR) at LANSCE

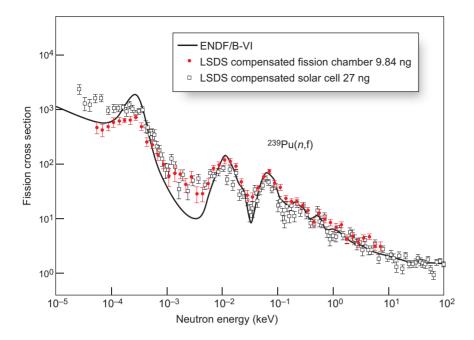


Figure 14. Measuring the Plutonium Fission Cross Section from a 9.87-Nanogram Sample at the LSDS

The measured fission cross sections of plutonium-239 from a 9.87-ng sample of plutonium-239 are in good agreement with the evaluated data, which are based on many other measurements and which are broadened here by the experimental energy resolution of the LSDS. These results demonstrate the capability of the LSDS for measuring the fission cross section of the isomeric state of uranium-235.

 $^{235}\text{U}$  is produced in the  $\alpha$ -decay of  $^{239}\text{Pu}$ 

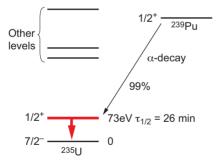


Figure 15. Energy Levels for the  $\alpha$ -Decay of Plutonium-239 Uranium-235m is produced in the decay of plutonium-239. The challenge is to separate a few nanograms of uranium from grams of plutonium by chemical means in a short time, before the isomer decays. The half-life of uranium-235m is only 26 min.

interact with the tungsten to produce spallation neutrons. Neutrons scatter in the lead and lose energy slowly, hence "slowing-down" in the name of this spectrometer. Samples and detectors are placed in channels in the lead assembly. We measure reaction rates, especially of neutron-induced fission, as a function of time and therefore of neutron energy—hence the term "spectrometer." The advantage of this approach is that the neutron flux on a sample is about 1000 times higher than in traditional beam-target experiments (refer back to Figure 2).

Whereas GEANIE and FIGARO require samples of several grams, and DANCE requires samples of a milligram or so, the LSDS can use samples in the nanogram (1 billionth of a gram) range. Measurements of very short-lived isotopes must be done with such small samples because of the intrinsic radioactivity and because often only very small amounts of the materials are available. Consequently, the incident-neutron flux must be much higher to get a measurable signal.

#### Addressing the Isomer Problem.

An uncertainty regarding the fission performance of a nuclear weapon is the fission of a 26-minute excited state of the uranium-235 nucleus, which can be produced in intense neutron fluxes. Recent theory suggests that the fission rate for this isomeric state of uranium-235 might be up to a factor of 2 less than that for the ground-state (unexcited) nucleus if the incident-neutron energies are low (below 200 keV). In contrast, older theories predicted that the cross section of the isomer might be much higher than that of the ground state. Despite the importance of knowing the relative fission cross sections in this energy range, an experimental measurement has never been made because it is very difficult to make enough of the isomer and complete the measurement in the short time that this state survives before decaying away.

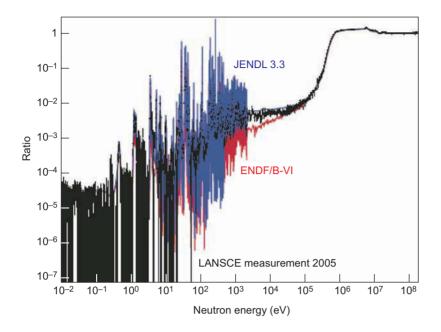


Figure 16. Ratio of Neptunium-237 to Uranium-235 Fission Cross Sections Note the discrepancy between the data and the values from the ENDF/B-VI file (accepted for use in the United States) in the range from 3000 to 100,000 eV. The ENDF/B-VI file is being updated to correct inaccuracies in this energy range. The JENDL 3.3 values are from the Japanese evaluated data library.

In the past few years, we and our colleagues from the CEA in France (the CEA supplied the lead assembly), the Chemistry Division at Los Alamos, and the Rensselaer Polytechnic Institute have developed the LSDS to measure the fission cross section of the isomeric state of uranium-235 and of other nuclides or isomers that can only be obtained in ultrasmall quantities. This measurement will not only provide data for applications but will be a unique probe into the fission physics of excited states of nuclei.

We have tested the assembly by measuring the well-known fission cross section for plutonium-239 on a very small sample, weighing only 9.8 nanograms (Figure 14). The results were accurate enough to enable plans for measuring the fission cross section of the isomeric state of uranium-235.

This short-lived, excited state of uranium-235 can be isolated by chemical techniques developed in the Los

Alamos Chemistry Division. The process involves taking a sample of plutonium-239 and letting it decay, which it does by alpha-particle emission.

Nearly all the decays (99%) populate the isomeric state of uranium-235 (Figure 15). The half-lives of the plutonium-239 parent and the isomer tell us that the equilibrium concentration of the isomer in the plutonium is only 2 parts in a billion. Thus, extreme demands are placed on the separation chemistry, which should exceed 1 part in 10<sup>10</sup> to enable a clean experiment on the isomer.

For the experiment on the isomer, we will place the sample in the LSDS and measure the fission rate as a function of time to get the energy dependence of the fission cross section, and we will continue to measure that rate as the isomer decays to the ground state of uranium-235, where the cross section is well known. In this sense, the experiment is "self-normalizing," and we will report the ratio of the

isomer-to-ground-state fission cross sections.

Many other reactions can be investigated with the LSDS, including fission cross sections and reactions that produce an alpha particle. The latter are especially interesting for understanding the formation of the elements in stars and for refining nuclear reaction models in ranges that desperately need more experimental data.

#### **Fission**

Improved accuracy for fission cross sections is another significant activity at LANSCE. An advanced fission chamber is being developed to identify the individual fission products, their energies, and their emission angles relative to the incident-neutron direction.

Our present measurements are with a conventional chamber, but with advanced electronics to improve on the techniques used for many years at Los Alamos. An example of the data is given in Figure 16 for the fission cross section of neptunium-237, measured relative to the standard uranium-235 over the full range of incident-neutron energy from thermal energies (0.025 electron volt) to 200 MeV. This work is the beginning of a major program at LANSCE to restart precision measurements of fission and to make significant improvements in the accuracy now required by the weapons program.

#### New and Unusual Nuclei

Finally, the new Isotope Production Facility (IPF) at LANSCE and the Chemistry Division at Los Alamos are equipped to fabricate the radioactive nuclear targets that can reasonably be measured (see the article "Accelerator Radioisotopes Save Lives" on page 112). These

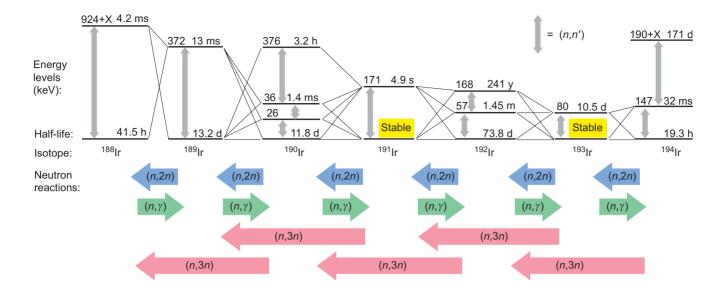


Figure 17. Realistic Network of Neutron-Induced Reactions on Iridium

This network of neutron-induced reactions is more realistic than the one shown in Figure 1. Here, the major isomeric (relatively long-lived) states are also shown as they need to be included in a comprehensive calculation. The energy of some isomeric levels is not known exactly—hence, the "+X" for two of the levels. Reactions on states living more than tens of days can, in principle, be studied with the instruments at LANSCE. The reaction rates on shorter-lived states must be calculated with the best physics models available.

isotopes are appropriate for experiments in our most-sensitive instruments. DANCE and the LSDS. Because the IPF is located on the same site as the nuclear data experiments, transportation of radioisotopes is easy and fast, and nuclei with rather short half-lives can be studied. Although this facility is dedicated to the production of medical radioisotopes, usable quantities of other radioactive isotopes may be produced at the IPF using its 100-MeV, 250-microampere proton beam and the facilities for processing irradiated targets in the Chemistry Division. We are presently working to prepare radioactive samples of arsenic-73 and gadolinium-153 that cause minimal disruption to the medical-radioisotope production program. These isotopes are important in assessing radiochemical diagnostic data from past nuclear tests.

Table II. Institutions in the LANSCE Nuclear Data Program

Instrument/Project	Institutions
GEANIE	Los Alamos National Laboratory (LANL), Lawrence Livermore National Laboratory (LLNL), Idaho National Laboratory (INL), Oak Ridge National Laboratory (ORNL), Commissariat à l'Énergie Atomique (CEA, France), and North Carolina State University (NC State)
FIGARO	LANL and CEA
DANCE	LANL, LLNL, ORNL, INL, Colorado School of Mines, NC State, and FZK Karlsruhe (Germany)
LSDS	LANL, LLNL, CEA, and Rensselaer Polytechnic Institute
Fission	LANL, Institute for Reference Materials and Measurements (European Union), LLNL, and INL
Others	Ohio University, Massachusetts Institute of Technology, University of Kentucky, Kyushu University (Japan), and Harvard University

## The Importance of Nuclear Physics

The capabilities described above are used to make direct measurements (or nearly direct) of the cross sections of importance to understanding nuclear-weapon performance. A more detailed look at the processes shows, however, that not all the reactions of possible importance can be investigated even with the significant advances in the LANSCE neutron sources and the associated instruments. Figure 1 above is a simplified picture of the reactions. Other isomers make the situation more complicated as shown in Figure 17, where 98 reactions are indicated (the thin lines and the arrows go in both directions). It will never be possible to measure all these reactions. Some are undoubtedly unimportant, but others may be extremely important. With the physics insights that come from the measurements that we can make, nuclear reaction models are put on a much firmer footing so that their predictions can assess the importance and magnitude of each of these reactions. With the new LANSCE capabilities, we can pin down the physics and the model parameters (optical model, nuclearlevel densities, gamma-ray strength functions, fission physics, and others) and extend our present knowledge into reactions on unstable nuclides and isomeric nuclear states.

## LANSCE—The Human Element

LANSCE has a dedicated staff of first-rate nuclear researchers who have developed the advanced neutron sources and innovative detectors described here in collaboration with colleagues from the Chemistry Division at Los Alamos and with coworkers from the U.S. and foreign laboratories and universities listed

in Table II. This array of human and instrumental assets produces crucial experimental data for use by nuclear theorists in the Applied Physics and Theoretical Physics Divisions at Los Alamos and the wider national and international nuclear physics community to deliver the most-essential nuclear data for making reliable predictions of weapon performance. Nuclear theory is an integral part of this effort because many of the relevant radioactive species have half-lives that are too short to measure in the laboratory even with our advanced instrumentation and the intense neutron sources at LANSCE. The continuing close collaboration among experimentalists, theorists, data evaluators, and applied scientists is essential to the significant advances made and expected in the interpretation of past nuclear-weapontest data and to the analysis and prediction of weapon performance in the absence of nuclear tests.

#### **Conclusions**

Nuclear data measurements for the nuclear weapons program are urgently needed to interpret nuclear test data for improving the calculational models so that they are applicable in a credible way to (1) quantify margins and uncertainties, (2) calculate effects caused by aging of materials and component replacement, and (3) investigate new designs, especially those that might be pursued by a rogue nation or terrorist organization. A steady investment in new types of sensitive detectors over the past few years has made possible relevant experimental measurements that were heretofore impossible. A bright future, with more innovative instruments, lies ahead.

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